

DOCKET NO.: 204698US0X

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF: : GROUP ART UNIT:  
DOMENICO SANFILIPPO ET AL :  
SERIAL NO.: NEW APPLICATION : EXAMINER:  
FILED: HEREWITH :  
FOR: PROCESS FOR THE PRODUCTION OF  
HYDROGEN

PRELIMINARY AMENDMENT

ASSISTANT COMMISSIONER FOR PATENTS  
WASHINGTON, D.C. 20231

SIR:

Prior to examination on its merits, please amend the above-identified application as follows:

IN THE CLAIMS

Please amend Claims 1-25.

--1. (Amended) A process for the production of hydrogen comprising: subjecting a solid to oxidation and treating the oxidized form thus produced with a reducing stream, in a different zone.

2. (Amended) The process for the production of hydrogen according to claim 1, comprising:

a) oxidation of a solid in a first reaction zone;

b) passage of the oxidized form of the solid to a reaction zone into which a reducing stream is fed, and its reaction with a hydrocarbon;

c) recovery of the reduced form of the solid and its feeding to the first reaction zone;

d) heat supply.

3. (Amended) The process of claim 2, wherein the heat supply is preferably effected during one of the two operations (b) and (c).

4. (Amended) The process of claim 2, wherein the solid, in the first reaction zone, is reacted with an agent selected from the group consisting of  $H_2O$ ,  $CO_2$ , and mixtures thereof.

5. (Amended) The process of claim 4, wherein the solid, in the first reaction zone, is preferably reacted with  $H_2O$ .

6. (Amended) The process of claim 2, wherein the solid subjected to oxidation in the first reaction zone comprises at least one element characterized by at least two different oxidation states, stable under the reaction conditions.

7. (Amended) The process of claim 6, wherein the solid, in the two different situations, is further characterized by different amounts of oxygen and enthalpy and is capable of cyclically and continuously passing from the reduced form to the oxidized form, and vice versa.

8. (Amended) The process of claim 7, wherein at least one redox element is present in the solid as binary compound corresponding to the formula



wherein Me is selected from Ce, Fe, W, Ni;

or as compounds corresponding to the formula



wherein Me is one or more elements selected from:

Ce, Pr, Co, Ni, Fe, Mo and W,

Z is one or more elements selected from Ce, Zr, V and Mo;

$x \geq 1$ ,  $y \geq 0$  and  $z \geq 1$ .

9. (Amended) The process for claim 8, wherein Me is equal to Fe.

10. (Amended) The process of claim 9, wherein the iron is present in the solid in a quantity ranging from 20 to 60% by weight.

11. (Amended) The process of claim 10, wherein Fe is present in the solid as a binary compound together with the binary compound of cerium and/or compounds corresponding to formula (8) wherein Me = Fe and Z = Ce.

12. (Amended) The process of claim 11, wherein the compound corresponding to formula (8) is  $\text{CeFeO}_3$ .

13. (Amended) The process of Claim 9, wherein the solid also contains a metal as promoter selected from Pt, Pd, Au and Rh.

14. (Amended) The process of claim 13, wherein the promoter is in a percentage ranging from 0.01 to 2% by weight.

15. (Amended) The process of Claim 9, wherein the solid also contains a transition metal as promoter selected from Cr, Mn, Nb and V.

16. (Amended) The process of claim 15, wherein the promoter is in a quantity ranging from 0.1 to 15% by weight.

17. (Amended) The process of Claim 12, wherein chromium is present as promoter.

18. (Amended) The process of Claim 8, wherein the reactive phase thus obtained can be used as such, or dispersed or supported on compounds including silica, alumina, and other

pure oxides including magnesium, calcium, cerium, zirconium, titanium, lanthanum, and mixtures thereof.

19. (Amended) The process of Claim 8, wherein the reactive phase is present in a quantity ranging from 20 to 80% by weight with respect to the compound which forms the carrier or the dispersing phase.

20. (Amended) The process wherein the reducing stream is selected from hydrocarbons, preferably aliphatic.

21. (Amended) The process of claim 20, wherein the aliphatic hydrocarbon is  $\text{CH}_4$ .

22. (Amended) The process of claim 2, wherein the heat supply takes place by the use of a supplementary thermal support unit, situated between the two reaction zones.

23. (Amended) The process of claim 22, wherein the heat supply is obtained using hydrogen as fuel.

24. (Amended) The process of claim 2, wherein the heat supply is obtained using methane or natural gas as fuel.

25. (Amended) The process of claim 1, characterized in that it comprises the following operations:

- oxidation of a solid in a first reaction zone; production of  $\text{H}_2$  or CO depending on the oxidizing compound;
- passage of the oxidized form of the solid to a subsequent reaction zone in which the reduction of the solid takes place by its reaction with a hydrocarbon;
- recovery of the reduced form of the solid and its feeding to the subsequent reaction zone;
- sending of the gaseous phase produced during the reduction of the solid to a suitable

separation section which allows the more or less complete separation of the complete combustion products ( $\text{CO}_2$  and  $\text{H}_2\text{O}$ ) from any possible non-converted hydrocarbon and from any possible by-products formed;

- possible recycling of the above gaseous stream to the reaction zone in which the reduction of the oxide takes place and/or to a further reaction zone, to enable complete conversion of the above stream to complete combustion products ( $\text{CO}_2$  and  $\text{H}_2\text{O}$ );

- elimination from the cycle of the complete combustion products ( $\text{CO}_2$  and  $\text{H}_2\text{O}$ ) coming from the purification section.

#### REMARKS

Claims 1-25 remain active in the case. They have been rewritten to improve grammar and usage without changing their scope.

The application is now believed to be in proper condition for examination on the merits and early notification of such action is earnestly solicited.

Respectfully submitted,

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**Marked-Up Copy**

Serial No: New Application

Preliminary Amendment Filed on:

March 19, 2001

--1. (Amended) A process for the production of hydrogen comprising: [consisting in] subjecting a solid to oxidation and treating the oxidized form thus produced with a reducing stream, in a different zone.

2. (Amended) The process for the production of hydrogen according to claim 1, comprising: [the previous claim characterized in that it comprises the following operations]:

- a) oxidation of a solid in a first reaction zone;
- b) passage of the oxidized form of the solid to a reaction zone into which a reducing stream is fed, and its reaction with a hydrocarbon;
- c) recovery of the reduced form of the solid and its feeding to the first reaction zone;
- d) heat supply.

3. (Amended) The process [for the production of hydrogen according to the previous] of claim 2, wherein the heat supply is preferably effected during one of the two operations (b) and (c).

4. (Amended) The process [for the production of hydrogen according to] of claim 2, [characterized in that] wherein the solid, in the first reaction zone, is reacted with an agent selected from the group consisting of  $H_2O$ , [and]  $CO_2$ , [or] and mixtures thereof [of the two].

5. (Amended) The process [for the production of hydrogen according to the previous] of claim 4, [characterized in that] wherein the solid, in the first reaction zone, is

preferably reacted with H<sub>2</sub>O.

6. (Amended) The process [for the production of hydrogen according to] of claim 2, [characterized in that] wherein the solid subjected to oxidation in the first reaction zone comprises at least one element characterized by at least two different oxidation states, stable under the reaction conditions.

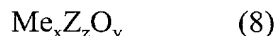
7. (Amended) The process of claim 6, wherein [for the production of hydrogen according to the previous claim, characterized in that] the solid, in the two different situations, is further characterized by different amounts of oxygen and enthalpy and is capable of cyclically and continuously passing from the reduced form to the oxidized form, and vice versa.

8. (Amended) The process of claim 7 [for the production of hydrogen according to the previous claim], wherein at least one redox element is present in the solid as binary compound corresponding to the formula



wherein Me is selected from Ce, Fe, W, Ni;

or as compounds corresponding to the formula



wherein Me is one or more elements selected from:

Ce, Pr, Co, Ni, Fe, Mo and W,

Z is one or more elements selected from Ce, Zr, V and Mo;

$x \geq 1$ ,  $y \geq 0$  and  $z \geq 1$ .

9. (Amended) The process [for the production of hydrogen according to] for claim 8,



wherein Me is equal to Fe.

10. (Amended) The process [according to] of claim 9, wherein the iron is present in the solid in a quantity ranging from 20 to 60% by weight.

11. (Amended) The process [for the production of hydrogen according to] of claim 10, wherein Fe is present in the solid as a binary compound together with the binary compound of cerium and/or compounds corresponding to formula (8) wherein Me = Fe and Z = Ce.

12. (Amended) The process [for the production of hydrogen according to] of claim 11, wherein the compound corresponding to formula (8) is  $\text{CeFeO}_3$ .

13. (Amended) The process of Claim 9 [for the production of hydrogen according to at least one of the claims from 9 to 12], wherein the solid also contains a metal as promoter selected from Pt, Pd, Au and Rh.

14. (Amended) The process [according to] of claim 13, wherein the promoter is in a percentage ranging from 0.01 to 2% by weight.

15. (Amended) The process of Claim 9 [for the production of hydrogen according to at least one of the claims from 9 to 14], wherein the solid also contains a transition metal as promoter selected from Cr, Mn, Nb and V.

16. (Amended) The process [according to] of claim 15, wherein the promoter is in a quantity ranging from 0.1 to 15% by weight.

17. (Amended) The process of Claim 12 [for the production of hydrogen according to claims 12 and 16], wherein chromium is present as promoter.

18. (Amended) The process of Claim 8 [for the production of hydrogen according to claims 8 to 17], wherein the reactive phase thus obtained [in turn] can be used as such, or

[suitably] dispersed or supported on compounds [such as] including silica, alumina, [or] and other pure oxides including [such as those of] magnesium, calcium, cerium, zirconium, titanium, lanthanum, and mixtures thereof [but also mixtures of these].

19. (Amended) The process of Claim 8, [for the production of hydrogen according to claims 8 to 18], wherein the reactive phase is present in a quantity ranging from 20 to 80% by weight with respect to the compound which forms the carrier or the dispersing phase.

20. (Amended) The process [for the production of hydrogen according to] of claim 2, [characterized in that] wherein the reducing stream is selected from hydrocarbons, preferably aliphatic.

21. (Amended) The process [for the production of hydrogen according to] of claim 20, wherein the aliphatic hydrocarbon is CH<sub>4</sub>.

22. (Amended) The process [for the production of hydrogen according to] of claim 2, wherein the heat supply takes place by the use of a supplementary thermal support unit, situated between the two reaction zones.

23. (Amended) The process [for the production of hydrogen according to the previous] of claim 22, wherein the heat supply is obtained using hydrogen as fuel.

24. (Amended) The process of [for the production of hydrogen according to] claim 2, wherein the heat supply is obtained using methane or natural gas as fuel.

25. (Amended) The process [for the production of hydrogen according to] of claim 1, characterized in that it comprises the following operations:

- oxidation of a solid in a first reaction zone; production of H<sub>2</sub> or CO depending on the oxidizing compound;
- passage of the oxidized form of the solid to a subsequent reaction zone in which the

reduction of the solid takes place by its reaction with a hydrocarbon;

- recovery of the reduced form of the solid and its feeding to the subsequent reaction

zone;

- sending of the gaseous phase produced during the reduction of the solid to a suitable separation section which allows the more or less complete separation of the complete combustion products ( $\text{CO}_2$  and  $\text{H}_2\text{O}$ ) from any possible non-converted hydrocarbon and from any possible by-products formed;

- possible recycling of the above gaseous stream to the reaction zone in which the reduction of the oxide takes place and/or to a further reaction zone, to enable complete conversion of the above stream to complete combustion products ( $\text{CO}_2$  and  $\text{H}_2\text{O}$ );

- elimination from the cycle of the complete combustion products ( $\text{CO}_2$  and  $\text{H}_2\text{O}$ ) coming from the purification section.--